

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>5</sup> :</b>  D06L 3/06	<b>A1</b>	<b>(11) International Publication Number:</b> WO 90/11400  <b>(43) International Publication Date:</b> 4 October 1990 (04.10.90)
<b>(21) International Application Number:</b> PCT/US90/01489 <b>(22) International Filing Date:</b> 20 March 1990 (20.03.90)  <b>(30) Priority data:</b> 325,718                      20 March 1989 (20.03.89)                      US  <b>(71) Applicant:</b> OLIN CORPORATION [US/US]; 350 Knotter Drive, P.O. Box 586, Cheshire, CT 06410-0586 (US).  <b>(72) Inventors:</b> GREEN, Donald, L. ; 12821 Valencia Way, Garden Grove, CA 92641 (US). GRYNKEWICH, Gregory, S. ; 2138 Ridgecrest Drive, Boise, ID 83712 (US). ALEXANDER, Roy, P. ; 23 Route #81, Killingsworth, CT 06417 (US).		<b>(74) Agents:</b> HAGLIND, James et al.; Olin Corporation, 350 Knotter Drive, Cheshire, CT 06410-0586 (US).  <b>(81) Designated States:</b> AT (European patent), AU, BB, BE (European patent), BF (OAPI patent), BG, BJ (OAPI patent), BR, CA, CF (OAPI patent), CG (OAPI patent), CH (European patent), CM (OAPI patent), DE (European patent), DK (European patent), ES (European patent), FI, FR (European patent), GA (OAPI patent), GB (European patent), HU, IT (European patent), JP, KP, KR, LK, LU (European patent), MC, MG, ML (OAPI patent), MR (OAPI patent), MW, NL (European patent), NO, RO, SD, SE (European patent), SN (OAPI patent), SU, TD (OAPI patent), TG (OAPI patent).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> ALKALI METAL CHLOROISOCYANURATE COMPOSITIONS  <b>(57) Abstract</b>  A composition in compressed form which is characterized by an alkali metal dichloroisocyanurate and a selected amount of a binding agent characterized by an alkali salt of an aliphatic carboxylic acid having from about 10 to about 20 carbon atoms and a salt of an alkyl ester of sulfuric acid, wherein the alkyl group contains from about 10 to about 18 carbon atoms. The binding agent provides the sanitizing tablet, in the presence of moisture, with increased integrity, reduced foaming or turbidity and substantially reduced residue formation.		

### DESIGNATIONS OF "DE"

Until further notice, any designation of "DE" in any international application whose international filing date is prior to October 3, 1990, shall have effect in the territory of the Federal Republic of Germany with the exception of the territory of the former German Democratic Republic.

#### *FOR THE PURPOSES OF INFORMATION ONLY*

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	ES	Spain	MG	Madagascar
AU	Australia	FI	Finland	ML	Mali
BB	Barbados	FR	France	MR	Mauritania
BE	Belgium	GA	Gabon	MW	Malawi
BF	Burkina Faso	GB	United Kingdom	NL	Netherlands
BG	Bulgaria	HU	Hungary	NO	Norway
BJ	Benin	IT	Italy	RO	Romania
BR	Brazil	JP	Japan	SD	Sudan
CA	Canada	KP	Democratic People's Republic of Korea	SE	Sweden
CF	Central African Republic	KR	Republic of Korea	SN	Senegal
CG	Congo	LI	Liechtenstein	SU	Soviet Union
CH	Switzerland	LK	Sri Lanka	TD	Chad
CM	Cameroon	LU	Luxembourg	TG	Togo
DE	Germany, Federal Republic of			US	United States of America
DK	Denmark	MC	Monaco		

-1-

## ALKALI METAL CHLOROISOCYANURATE COMPOSITIONS

This invention relates to compressed forms of alkali metal salts of chlorinated isocyanuric acid and to a process for producing the same. More particularly, the invention relates to improved bleaching and water sanitizing compositions having as the active ingredient an alkali salt of dichloroisocyanuric acid, and to the preparation of such compositions.

The solid salts of chlorinated isocyanuric acid, such as the alkali metal dichloroisocyanurates, are well-known sources of available chlorine. They are used in a variety of bleaching and sanitizing applications. Depending on the type of physical product desired for such use, these salts can be provided in granular or compressed form, such as tablets, sticks and the like. The compressed form is often preferred inasmuch as it would enable the slow release of chlorine at reasonably controlled rates. Other reasons for preferring the compressed or tabletted form of these sanitizing products include ease or simplicity of packaging and consumer appeal.

A number of developments have been made relating to the production of tabletted forms of chlorinated isocyanuric acid and salts thereof. Thus, U.S. Patent No. 3,325,411 to F. N. Stepanek, Jr. describes a method for tableting such products; and U.S.

-2-

Patent No. 3,342,674 to X. Kowalski discloses an improvement in this art wherein a salt of an aliphatic carboxylic acid having at least 10 carbon atoms is used as a lubricant to facilitate the tableting operation.

5           The desirability of compressed forms of alkali metal salts of dichloroisocyanuric acid for use in industrial and institutional sanitizing applications has been recognized and sought after in recent years. Large tablets, sticks or discs of such salts, placed in a  
10   suitable holder/dispenser, would provide a steady source of available chlorine for a prolonged period of time, thereby eliminating the need for frequent replenishment of the sanitizer. Such a product has been particularly sought after for use in such applications as  
15   institutional and industrial laundry and dishwashing machines.

          However, when tablets of these salts are exposed for prolonged periods of time to a wet environment, e.g., repeated exposure to a water spray as  
20   used in dish or laundry washing machines, the tablet integrity suffers significantly, resulting in an undesirably rapid or erratic delivery of chlorine.

          Tablet integrity can be improved by the use of alkali metal salts of stearic acid i.e., sodium  
25   stearate. The use, however, of these compounds having low water solubility results in the deposition of a highly undesirable, water-insoluble residue on items washed or machine parts and surfaces.

          As these compressed forms are used in laundry  
30   or dishwashing machines in which high rates of agitation occur, it is also desirable to prevent the formation of significant amounts of foam.

-3-

Brazilian Patent No. PI 85/794 issued to A. E. Milford et al teaches the use of alkali salts of fatty sulfates as lubricants in the production of compressed forms of chloroisocyanuric compounds. This lubricant, however, results in the formation of excessive amounts of foam.

Thus there is a need for water soluble tablets containing an alkali metal salt of dichloroisocyanuric acid having improved tablet integrity while minimizing the deposition of water-insoluble residues.

Further, there is a need for water soluble tablets containing an alkali metal salt of dichloroisocyanuric acid which form only limited amounts of foam when used in a laundry or dishwashing application.

Now an improved composition of alkali metal dichloroisocyanurate in compressed form has been found which overcomes these problems. In accordance with this invention, the composition is comprised of an alkali metal dichloroisocyanurate and a selected amount of a binding agent comprised of an alkali salt of an aliphatic carboxylic acid having from about 10 to about 20 carbon atoms, and a salt of an alkyl ester of sulfuric acid, wherein the alkyl group contains from about 10 to about 18 carbon atoms.

The use of this binding agent in making compressed forms of an alkali metal dichloroisocyanurate has been found to be effective in stabilizing the integrity of the tablet in a wet environment while at the same time minimizing or eliminating the formation of residue and foam.

More in detail, any alkali metal salt of dichloroisocyanuric acid and its hydrates may be used in preparing the composition of the invention. However, for practical reasons the sodium and potassium salts are preferred.

-4-

The binding agent of the invention has as one component an alkali salt of an aliphatic carboxylic acid having from about 10 to about 20 carbon atoms. Suitable for use are alkali salts of aliphatic monocarboxylic acids including decanoic (caproic) acid, undecanoic (hendecanoic) acid, dodecanoic (lauric) acid, tridecanoic acid, tetradecanoic (myristic) acid, pentadecanoic acid, hexadecanoic acid, octadecanoic (stearic) acid, icosanoic acid, and mixtures thereof. The alkali salts of these acids which are employed include those of alkali metals such as sodium, potassium, and lithium, and alkaline earth metals, for example, calcium and magnesium, as well as mixtures of alkali metal and alkaline earth metal salts.

Preferred are aliphatic carboxylic acids having from about 12 to about 18 carbon atoms, including mixtures thereof, where the alkali salt is an alkali metal salt.

More preferred are alkali metal salts of hexadecanoic acid and octadecanoic (stearic acid) including potassium hexadecanoate and sodium stearate.

The second essential component of the binding agent of the novel composition of the invention is a salt of an alkyl ester of sulfuric acid where the alkyl group has from about 10 to about 18 carbon atoms. Salts which can be employed are those of the alkali metals such as sodium, potassium, or lithium, the ammonium salts, the alkaline earth metals such as magnesium or calcium as well as mixtures thereof. Alkyl groups having from about 10 to about 18 carbon atoms which may be used include decyl (capryl), undecyl (hendecyl), dodecyl (lauryl), tridecyl, tetradecyl (myristyl), hexadecyl (cetyl), octadecyl (stearyl) groups, and mixtures thereof.

-5-

Preferred embodiments are sodium and magnesium salts of alkyl esters of sulfuric acid where the alkyl group has carbon atoms in the range of from about 10 to about 14, including mixtures thereof, with sodium lauryl sulfate  
5 and magnesium lauryl sulfate being particularly preferred.

In the binding agent of the present invention, the alkali salt of an aliphatic carboxylic acid and the salt of an alkyl ester of sulfuric acid are used in amounts which provide, for example, a weight ratio of  
10 alkali salt of the aliphatic carboxylic acid to the salt of the alkyl ester of sulfuric acid of from about 3:1 to about 1:5. Preferred embodiments contain a weight ratio of alkali salt of the aliphatic carboxylic acid to the salt of the alkyl ester of from about 2:1 to about 1:3,  
15 and more preferably, from about 1.5:1 to about 1:2.

The novel compositions of the present invention contain selected amounts of the binding agent including up to about 5 percent by weight. Compressed compositions of the present invention produced at  
20 commonly used compaction pressures preferably include from about 0.25 to about 3.5 percent by weight, and more preferably from about 0.5 to about 2.5 percent by weight of the compressed composition.

These compositions release effective amounts  
25 of the alkali metal dichloroisocyanurate sanitizing agent for use in dish or laundry washing applications. Surprisingly, the binding agent comprised of the alkali salt of an aliphatic carboxylic acid and the salt of an alkyl ester of sulfuric acid provides the sanitizing  
30 tablet, in the presence of moisture, with increased integrity, reduced foaming or turbidity and substantially reduced residue formation.

-6-

Compositions of the present invention are compressed into tablets using known methods of tableting and apparatus therefore. Other compressed forms of the compositions such as briquets, sticks, cylinders, blocks,  
5 or pellets may also be produced.

The compositions of the present invention are further illustrated by the following examples with no intention of being limited thereby. All percentages are by weight unless otherwise specified.



-7-

EXAMPLE 1

A mixture of sodium dichloroisocyanurate dihydrate containing sodium lauryl sulfate (0.5 percent) and sodium stearate (0.5 percent) was prepared. The  
5 mixture was pressed in a 3 inch diameter tabletting die at 25 tons pressure to produce tablets containing about 200 grams. The top and side of the tablet were shrink-wrapped with polyvinylchloride film and the tablet placed on an open mesh rack with the exposed face in  
10 contact with the rack. A spray nozzle, positioned below the rack, sprayed water (130°F, 54°C) for a period of 30 seconds. The available chlorine-containing solution was collected in a container and the height of any foam produced was measured. The solution was then decanted  
15 into a volumetric flask until the solution height was about half-way up the neck. The solution was allowed to stand until any insoluble scum present floated to the top. The thickness of the scum layer was measured, along with the available chlorine concentration and the  
20 turbidity of the solution. The water spraying was repeated at intervals until the tablet dissolved. The results are recorded in Table I below.

Comparative Examples A, B, and C

The procedure of Example 1 was followed  
25 exactly using sodium dichloroisocyanurate without an additive (Example A); a mixture of sodium dichloroisocyanurate dihydrate and sodium stearate (1 percent)(Example B); and a mixture of sodium dichloroisocyanurate and sodium lauryl sulfate (1  
30 percent) (Example C). The results are given in Table I below.

TABLE I

Example	Time	Scum Height (Inches)	Solution Weight (Grams)	AvCl (%)	Turbidity (NTU)	Foam Height (Inches)
1	09:39 AM	0.08	1113		80	0.00
	09:53 AM	0.07	927	0.55	68	0.00
	10:11 AM	0.08	921	0.50	56	0.00
	10:23 AM	0.10	927	0.54	71	0.00
	10:33 AM	0.08	934	0.55	59	0.00
	11:07 AM	0.05	940	0.44	60	0.00
	11:24 AM	0.12	957	0.58	70	0.00
	11:43 AM	0.10	934	0.67	78	0.00
	12:01 PM	0.08	907	0.60	70	0.00
	12:18 PM	0.13	938	0.73	75	0.00
	12:43 PM	0.09	933	0.59	66	0.00
	01:03 PM	0.15	952	0.78	88	0.00
	01:18 PM	0.12	932	0.79	92	0.00
	01:31 PM	0.13	945	0.90	99	0.00
	01:44 PM	0.15	945	0.95	94	0.00
	01:58 PM	0.17	958	1.09	110	0.00(1)

(1) Tablet 1 inch in diameter broke up and was mushy.

TABLE I (Cont'd.)

Comparative Example A	Time	Scum Height (Inches)	Solution Weight (Grams)	AvCl (%)	Turbidity (NTU)	Foam Height (Inches)
	09:55 AM	0.00	882	2.20	8.6	0.00
	10:15 AM	0.00	855	0.94	8.2	0.00
	10:33 AM	0.00	893	0.91	7.3	0.00
	10:48 AM	0.00	896	1.00	8.1	0.00
	11:04 AM	0.00	890	1.24	9.0	0.00
	11:23 AM	0.00	909	1.75	10.2	0.00
	11:50 AM	0.00	902	1.66	12.1	0.00
	12:26 PM	0.00	942	1.43	10.7	0.00
	12:46 PM	0.00	940	0.77	6.3	0.00 (2) <sub>p</sub>

(2) End of tablet.

TABLE I (Cont'd.)

Comparative Example B	Time	Scum Height (Inches)	Solution Weight (Grams)	AvCl (%)	Turbidity (NTU)	Foam Height (Inches)
	01:05 PM	0.55	921	0.59	117	0.00
	01:24 PM	0.48	939	0.59	95	0.00
	01:45 PM	0.35	930	0.47	117	0.00
	02:07 PM	0.35	930	0.61	90	0.00
	02:29 PM	0.37	926	0.52	87	0.00
	02:47 PM	0.38	923	0.59	74	0.00
	03:07 PM	0.35	930	0.60	160	0.00
	03:26 PM	0.42	930	0.70	130	0.00
	03:40 PM	0.48	942	0.88	138	0.00
	03:55 PM	0.65	959	1.52	162	0.00
	04:08 PM	0.88	935	1.46	190	0.00(3)

-10-

(3) Tablet mushy, about 2" D. and .2" H.

TABLE I (Cont'd.)

Time	Scum Height (Inches)	Solution Weight (Grams)	AvCl (%)	Turbidity (NTU)	Foam Height (Inches)
Comparative					
Example C					
10:38 AM	0.00	887	0.37	5	0.00
10:55 AM	0.00	917	0.38	7	0.00
11:10 AM	0.00	948	0.40	8	0.20
11:28 AM	0.00	950	0.39	7	0.32
11:43 AM	0.00	930	0.45	8	0.38
12:00 PM	0.00	949	1.20	6	0.40
12:15 PM	0.00	951	0.51	6	0.56
12:28 PM	0.00	945	0.49	7	0.48
12:42 PM	0.00	955	0.58	7	0.50
12:53 PM	0.00	957	0.64	6	0.65
01:08 PM	0.00	960	0.63	6	0.58
01:25 PM	0.00	967	0.69	6	0.60
01:41 PM	0.00	972	0.93	7	0.75
01:56 PM	0.00	986	1.52	12	1.00
02:15 PM	0.00	982	1.41	11	0.95(4)

-11-

(4) Tablet was soft, mushy, and completely dissolved on one side.

-12-

TABLE I (Cont'd.)

<u>Summary</u>	<u>Average Scum</u>	<u>Average Turbidity</u>	<u>Average Foam</u>	<u>Average AvCl</u>
Example 1	0.11	77.25	0.00	0.68
5 Comparative Example A	0.00	8.94	0.00	1.32
Comparative Example B	0.48	123.64	0.00	0.77
10 Comparative Example C	0.00	7.31	0.49	0.71

The tablet of Example 1 showed high integrity, no foam formation, an average of 77 percent reduction in scum formation, and consistent release of available chlorine concentration over the tablets of Comparative

15 Examples A, B, and C.

EXAMPLES 2-7

Mixtures of sodium dichloroisocyanurate (1.96-1.98 grams) with varying amounts of sodium lauryl sulfate and sodium stearate were made. Each mixture was

20 poured into a 1.6 cm diameter tabletting die and the die placed in a hydraulic Carver press and pressed into a tablet at one of three different pressures. Filter paper (Whatman #1) was placed on the domed bottom of a

25 container, the top of the dome being about 0.25 cm above the bottom of the container. The filter paper was wetted with 3.5 grams of water with the excess water settling at the base of the dome. The tablet was placed at the highest point of the dome of the wetted filter paper so

-13-

that it contacted the wetted filter paper but not the excess water. As the tablet absorbed water it was converted in time from a hard brittle solid to a soft mushy paste. The disintegration point was evidenced by a darkening in color of the composition and the change in physical state was confirmed by tapping with a metal spatula. At this point, the time was recorded. The tablet compositions, tableting pressures and disintegration times are recorded in Table II below.

10

Comparative Example D-J

The procedure of Examples 2-7 was followed exactly with the exception that sodium lauryl sulfate or sodium stearate or both sodium lauryl sulfate and sodium stearate were omitted from the mixture. The results are given in Table II below.

-14-

TABLE IIDisintegration Points of Tablets

	Example No.	SDCC* (Grams)	SLS** (Grams)	NaSt! (Grams)	Tabletting Pressure (Pounds)	Disintegration Point (Minutes)
5	1	1.97	0.02	0.01	1300	1.75
	2	1.97	0.01	0.02	1300	6.70
	3	1.97	0.01	0.02	2050	6.63
	4	1.96	0.02	0.02	2050	9.08
10	5	1.98	0.05	0.05	2800	7.80
	6	1.96	0.02	0.02	2800	14.75
	7	1.97	0.02	0.01	2800	6.12
	Comp. D	2.00	0.00	0.00	1300	0.53
15	Comp. E	1.98	0.00	0.02	1300	3.93
	Comp. F	1.99	0.01	0.00	1300	1.08
	Comp. G	1.98	0.02	0.00	2050	1.27
20	Comp. H	2.00	0.00	0.00	2800	0.48
	Comp. I	1.98	0.00	0.02	2800	9.55
	Comp. J	1.98	0.01	0.00	2800	7.06

\*SDCC = sodium dichloroisocyanurate

\*\*SLS = sodium laurylsulfate

!NaSt = sodium stearate



-15-

The compositions of the present invention, as illustrated by Examples 2-7, provide significant increases in the disintegration points over those illustrated by the Comparative Examples.

-16-

WHAT IS CLAIMED IS:

1. A composition in compressed form which is characterized by an alkali metal dichloroisocyanurate and a selected amount of a binding agent characterized by an alkali salt of an aliphatic carboxylic acid having from about 10 to about 20 carbon atoms and a salt of an alkyl ester of sulfuric acid, wherein the alkyl group contains from about 10 to about 18 carbon atoms.
2. The composition of claim 1 characterized in that a weight ratio of the alkali salt of an aliphatic carboxylic acid to the salt of an alkyl ester of sulfuric acid is from about 3:1 to about 1:5.
3. The composition of claim 1 characterized in that a alkali salt of an aliphatic carboxylic acid is selected from the group consisting of alkali metals, alkaline earth metals, and mixtures thereof.
4. The composition of claim 1 characterized in that the salt of an alkyl ester of sulfuric acid is selected from the group consisting of alkali metals, alkaline earth metals, and mixtures thereof.
5. The composition of claim 1 characterized in that the compressed form is selected from the group consisting of tablets, briquets, sticks, cylinders, blocks, or pellets.
6. The composition of claim 1 characterized in that the alkali metal dichloroisocyanurate is selected from the group consisting of sodium dichloroisocyanurate, potassium dichloroisocyanurate, lithium dichloroisocyanurate, their hydrates, and mixtures thereof.

-17-

7. The composition of claim 6 characterized in that the binding agent is present in amounts up to about 5 percent by weight.

8. The composition of claim 6 characterized in that the alkali salt of an aliphatic carboxylic acid is an alkali metal salt of a monocarboxylic acid having from about 12 to about 18 carbon atoms.

9. The composition of claim 8 characterized in that the salt of an alkyl ester of sulfuric acid is an alkali metal salt and the alkyl group contains from about 10 to about 14 carbon atoms.

10. The composition of claim 9 characterized in that the alkali metal dichloroisocyanurate is sodium dichloroisocyanurate and hydrates thereof.

11. The composition of claim 10 characterized in that the weight ratio of alkali metal salt of monocarboxylic acid to the alkali metal salt of the alkyl ester of sulfuric acid is from about 2:1 to about 1:3.

12. The composition of claim 11 characterized in that the binding agent is present in amounts of from about 0.25 to about 3.5 percent by weight.

13. The composition of claim 10 characterized in that the alkali metal salt of a monocarboxylic acid is sodium.

14. The composition of claim 13 characterized in that alkyl ester of sulfuric acid contains alkyl groups having from about 10 to about 14 carbon atoms, and mixtures thereof.

15. The composition of claim 14 characterized in that the alkali metal salt of the alkyl ester of sulfuric acid is sodium.

16. The composition of claim 15 characterized in that the monocarboxylic acid is hexadecanoic acid, stearic acid, and mixtures thereof.

17. The composition of claim 16 characterized in that the weight ratio of sodium salt of monocarboxylic acid to the sodium salt of the alkyl ester of sulfuric acid is from about 1.5:1 to about 1:2.

18. The composition of claim 16 characterized in that the sodium salt of the monocarboxylic acid is sodium stearate.

19. The composition of claim 18 characterized in that the sodium salt of the alkyl ester of sulfuric acid is sodium lauryl sulfate.

20. The composition of claim 19 characterized in that the compressed form is a tablet.

# INTERNATIONAL SEARCH REPORT

International Application No. **PCT/US90/01489**

## I. CLASSIFICATION OF SUBJECT MATTER (In several classification symbols apply, indicate all) \*

According to International Patent Classification (IPC) or to both National Classification and IPC

**INT CL (5) D06L 3/06**  
**H.S. 252/186.250**

## II. FIELDS SEARCHED

Minimum Documentation Searched †

Classification System

Classification Symbols

**US**

**252/186.250**  
**252/187.340**

Documentation Searched other than Minimum Documentation  
to the Extent that such Documents are Included in the Fields Searched \*

## III. DOCUMENTS CONSIDERED TO BE RELEVANT \*

Category *	Citation of Document, † with indication, where appropriate, of the relevant passages ‡	Relevant to Claim No. ‡
<b>Y</b>	<b>US, A, 3,488,420 Published 06 January 1970 column 1 lines 23-27, column 2 line 41, column 3 lines 12-21</b>	<b>1-3,5-8,10,11</b>
<b>Y</b>	<b>US, A, 3,342,674 Published 19 September 1967 column 1, lines 1-15, 24-35, 60-63, column 2 lines 31-34, column 4 lines 35-46 column 5 lines 60-45, column 6 lines 1-6</b>	<b>1,2,5-8,10-13 16-18,20</b>
<b>Y</b>	<b>US, A, 3,325,411 Published 13 June 1967 column 1 lines 38-51, column 2 lines 12-19, 37-44 column 3 lines 28-35</b>	<b>1,2,4-6,7,9- 12,14,15,17, 19,20</b>

### \* Special categories of cited documents: †

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"Δ" document member of the same patent family

## IV. CERTIFICATION

Date of the Actual Completion of the International Search

Date of Making of this International Search Report

**06 JUNE 1990**

**11 JUL 1990**

International Searching Authority

**ISA/US**

Signature of Authorized Officer

**VALERIE D. FEE**

Form PCT/ISA/210 (second sheet) (Rev. 11-87)